Selective Dynamic Nuclear Spin Polarization in Spin-Blocked Double-Dot

Changxue Deng and Xuedong Hu

Department of Physics, University at Buffalo, SUNY, Buffalo, NY 14260-1500
(Dated: February 2, 2008)

We study the mechanism of dynamical nuclear spin polarization by hyperfine interaction in spin-blocked double quantum dot system. We calculate the hyperfine transition rates and solve the master equations for the nuclear spins. Specifically, we incorporate the effects of the nuclear quadrupole coupling due to the doping-induced local lattice distortion and strain. Our results show that nuclear quadrupole coupling induced by the 5% indium substitution can be used to explain the recent experimental observation of missing arsenic NMR signal in the spin-blocked double dots.

In many metals and semiconductors, nuclei and conduction electrons form a coupled nonlinear spin system through the hyperfine interaction. Such a coupling is well studied in bulk experimental phenomena such as dynamical nuclear spin polarization [1]. Both electron and nuclear spins in semiconductor heterostructures have been proposed as candidates for qubits in a scalable quantum information processor. There are also a variety of spintronic devices where electron spins provide novel functionalities. For both quantum computation and spintronics, understanding the electron-nuclear spin interaction and the resulting coupled dynamics is crucial because of its important roles in spin manipulation and decoherence. Indeed, in the past decade there have been extensive experimental and theoretical studies of electron-nuclei hyperfine interaction in semiconductor nanostructures [2, 3, 4, 5, 6]. In this Letter, we show that nuclear quadrupole coupling can be a critical factor in this coupled dynamics, thus needs to be carefully accounted for in the study of spin-based quantum computers and spintronic devices.

Dynamical nuclear spin polarization has recently been demonstrated in a spin blocked semiconductor double quantum dot made of $Ga_{0.95}In_{0.05}As$ [6, 7, 8]. Coulomb interaction and Pauli principle dictates that in a double quantum dot two-electron singlet and triplet states are split, so that proper voltage offset and bias between the double dot leads to significant suppression in the tunnel current due to occupied triplet state (thus the so-called spin blockade regime, which has been suggested for single spin detection, a crucial component of quantum information processing) [7]. One way to lift this blockade is to apply an appropriate magnetic field, so that electron-nuclear spin hyperfine coupling can facilitate electron spin-flip transitions, which in turn leads to dynamical nuclear spin polarization as was shown in Ref. [6]. The reported experimental observations reveal several quite unexpected phenomena [6], one of which is that while nuclear magnetic resonance (NMR) signals were observed at the ^{69}Ga and ^{71}Ga frequencies, no comparable response has been found from the ^{75}As nuclei. It seems that nuclear spin polarization cannot be built up in

^{75}As nuclei even though they have the largest concentration and the largest hyperfine coupling with the electrons among the three nuclear species.

In this Letter we study the mechanism of dynamical nuclear spin polarization by hyperfine interaction in a spin-blocked double quantum dot, taking into account electron tunneling and nuclear quadrupole coupling. We show that the absence of ^{75}As nuclear spin signal is due to the local strain created by the presence of Indium atoms. Specifically, the substitution of the 5% In for Ga breaks the crystal symmetry and induces a local lattice distortion [9, 10]. The charge redistribution generates *static* electric field gradients (EFG) at the surrounding lattice sites, which lead to finite quadrupole coupling for the nuclei [11]. The randomly distributed In atom only substitute one of the nearest neighbors of As atoms, while the nearest neighbors of Ga atoms are always four As atoms. Therefore the local electric field gradients at the Ga nuclei sites are on average much smaller than at the As sites. We show that this difference of EFG at the locations of As and Ga nuclei is the major origin of the unexpected absence of As nuclear spin signal.

We first note that the static quadrupole interaction alone cannot generate any transition among the spin eigenstates because there is no energy relaxation mechanism. This is different from the spin-lattice effect, where the EFG is generated by the lattice oscillations. There quadrupole spin relaxation can be realized by absorbing and emitting a phonon simultaneously (i.e., a Raman process) [12]. In the system we consider, the nuclei couple with the electrons, which in turn couple to the leads (reservoir) by tunneling, so that energy conservation can be satisfied through charge exchange between the dot and the leads. In the following we first find the nuclear spin eigenstates including the static quadrupole interaction, then evaluate the first-order transition rates among these eigenstates due to hyperfine coupling.

The static Zeeman and quadrupole Hamiltonian for a single nuclear spin is

$$H_n = -\omega \hat{I}_z + \nu(\hat{I}_Z^2 - \frac{1}{3}\hat{I}^2), \qquad (1)$$

where ω is the nuclear spin Lamour frequency and ν is quadrupole coupling strength that is proportional to the EFG at the locations of the nuclei, z is the direction of external magnetic field and Z is the principle axis of the largest EFG [13]. We have neglected the asymmetrical effect of quadrupole interaction. It was determined experimentally that $\nu_{As} \approx 2.5$ MHz and ν_{Ga} is much less than ν_{As} in $In_x Ga_{1-x} As$ for x up to 0.01 [10]. For a typical external field B_{ext} (0.8 Tesla) applied in the experiment [6], we can assume $\nu_{Ga} \ll \omega_{Ga} \approx 10.4$ and 8.2 MHz for ^{69}Ga and ^{71}Ga nuclei. Since our intention is to study the relation of As and Ga nuclear polarization, we will not differentiate ^{69}Ga and ^{71}Ga further in this Letter. The small ν_{Ga} allows eigenstates $|\phi_i\rangle$ (i=-3/2,-1/2,1/2 or 3/2) of H_n for Ga nuclei to be expressed perturbatively in terms of the Zeeman spin eigenstates $|m\rangle$.

$$|\phi_{\pm\frac{3}{2}}\rangle = |\pm\frac{3}{2}\rangle - b\,|\pm\frac{1}{2}\rangle \mp c\,|\mp\frac{1}{2}\rangle,$$

$$|\phi_{\pm\frac{1}{2}}\rangle = |\pm\frac{1}{2}\rangle + b\,|\pm\frac{3}{2}\rangle \mp c\,|\mp\frac{3}{2}\rangle, \qquad (2)$$

where $b = \sqrt{3}\nu_{Ga}\sin\theta\cos\theta/\omega_{Ga}$ and $c = \sqrt{3}\nu_{Ga}\sin^2\theta/4\omega_{Ga}$. θ is the angle between z and Z axes. In these calculations x axis is chosen to be in the plane of z and Z. These new states $|\phi_i\rangle$ are not eigenvectors of $\hat{I}_z$ anymore, though their average spin polarizations $\langle\phi_i|\hat{I}_z|\phi_i\rangle = i + O(\nu_{Ga}^2/\omega_{Ga}^2)$.

Transitions can be induced by the hyperfine interaction among the new energy eigenstates of the Ga nuclei. The hyperfine Hamiltonian is

$$H_h = \sum_i^N A_i |\psi(\mathbf{R}_i)|^2 \left[\hat{I}_z^i \hat{S}_z + \frac{1}{2}(\hat{I}_+^i \hat{S}_- + \hat{I}_-^i \hat{S}_+)\right]. \quad (3)$$

Here $N \sim 10^5$ is the number of nuclei in a quantum dot; A is the hyperfine coupling constant; $\psi(\mathbf{R}_i)$ is the electron envelope wavefunction at the ith nucleus. Using the electron density at nuclear sites [14] we estimate $A \approx 46$ μeV and 40 μeV for As and Ga nuclei respectively. Hamiltonian (3) describes the interaction of an electron with an ensemble of N nuclei in the dot.

To calculate the transition rates given in Fig. 1 we must study how the system couples to the environment to satisfy energy conservation. In the present case electron exchange between a dot and the leads is the most important energy relaxation mechanism [15]. For triplet state $|\downarrow\downarrow\rangle$ [16] the electron in the 1st (2nd) dot only couples to the left (right) lead. The electron tunnel Hamiltonian can be expressed as

$$H_e = \sum_k \epsilon_k c_{k\downarrow}^+ c_{k\downarrow} + \epsilon_0 d_\downarrow^+ d_\downarrow + \sum_k \left[t_k c_{k\downarrow}^+ d_\downarrow + \text{H.C.}\right], \quad (4)$$

where ϵ_0 is the renormalized single particle energy level. Since the Hamiltonian H_e is quadratic, exact solution of the retarded Green's function $G_{\downarrow\downarrow}^{Ret} =$
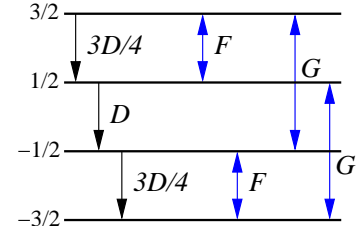

FIG. 1: Schematic of hyperfine induced transitions among the energy eigenstates of Hamiltonian H_n. The downward arrows represent simultaneous flip-flop scattering of nuclei and the electron in a triplet state. F and G are the nuclear spin relaxation rates induced by the effective hyperfine magnetic field of the electron ($\frac{A}{N}I_z S_z$).

$-i\theta(t)\langle\{d_\downarrow(t), d_\downarrow^+(0)\}\rangle$ can be obtained by using the equation of motion method. The density of state (DOS) follows from $\rho_T(\epsilon) = -\text{Im}G_{\downarrow\downarrow}^{Ret}(\epsilon+i0^+)/\pi$. It is straightforward to show that the electron triplet state is broadened

$$\rho_T(\epsilon) = \frac{1}{2\pi}\frac{\Gamma}{(\epsilon-\epsilon_0)^2 + \Gamma^2/4}, \quad (5)$$

where $\Gamma = 2\pi\sum_k |t_k|^2\delta(\epsilon-\epsilon_k)$ is the level broadening for the triplet state. For the electron in the singlet state, the DOS should be modified by a weak inter-dot coupling. However, the major energy relaxation channel is still the lead-dot tunnel coupling, so we can approximate the singlet DOS with Eq. (5) as well.

The first term in Eq. (3) does not involve electron spin flip. However, it can induce transitions among the energy eigenstates due to mixture of different spin eigenstates in Eq. (2). The transition rate between state $|\phi_i\rangle$ and $|\phi_j\rangle$ due to this term is given by the Fermi golden rule,

$$W_{i,j}^z = \frac{\pi A^2}{2\hbar N^2}|\langle\phi_i|\hat{I}_z|\phi_j\rangle|^2 P(E_j - E_i), \quad (6)$$

where E_i and E_j are the eigenenergies of H_n. The absorption power P is

$$P(\Delta) = \sum_{\epsilon_i,\epsilon_f} \rho(\epsilon_i)\rho(\epsilon_f)\delta(\epsilon_f - \epsilon_i + \Delta), \quad (7)$$

where $\rho(\epsilon)$ is the DOS of either the electron singlet or the triplet state. Using Eq. (6), the transition rates in Fig. (1) can be calculated: $F = 6b^2 A^2/(N^2\hbar\Gamma)$ and $G = 6c^2 A^2/(N^2\hbar\Gamma)$. In deriving these expressions we have neglected the nuclear energy difference of the initial and final state since they are much smaller than the width of electronic energy broadening.

The calculation of the transition rates due to the hyperfine flip-flop terms in Eq. (3), which correspond to the polarized triplet to singlet electron transitions, is more complicated. When the two electron states are degenerate, the hyperfine flip-flop transition is on resonance,

leading to nuclear spin polarization. However, the transition should quickly becomes off resonance due to the effective nuclear magnetic field B_N experienced by the electrons. The effective nuclear magnetic fields can be estimated as

$$B_N = \gamma_{As}\frac{\langle \hat{I}_z \rangle_{As}}{I} + \gamma_{Ga}\frac{\langle \hat{I}_z \rangle_{Ga}}{I}, \qquad (8)$$

where $\gamma_{As} \approx -2.8$ Tesla and $\gamma_{Ga} \approx -1.3$ Tesla [14]. As and Ga nuclei have independent spin temperatures because their mutual flip-flop process is largely suppressed due to different magnetic moments. Equilibrium in nuclear spin polarization will thus be established in each of the nuclear species independently. B_N can be quite large for even moderate nuclear polarization. The energy difference of the singlet state and triplet state due to B_N can be approximated by $\Delta_{ST} = -g^*\mu_B B_N$ assuming the energy of the singlet state is independent of magnetic field. The flip-flop transition rates of hyperfine interaction from state $|\phi_i\rangle$ to state $|\phi_j\rangle$ is thus

$$W_{i,j}^- = \frac{f_T \pi A^2}{8\hbar N^2}|\langle \phi_j|\hat{I}_-|\phi_i\rangle|^2 P(\Delta_{ST} + E_j - E_i), \qquad (9)$$

where f_T is a multiplication factor due to cotunneling and the probability of the triplet state occupation (electrons can tunnel into three triplet states and one singlet state, but only one triplet is relevant to nuclear polarization). f_T should be multiplied by another factor 0.5 because there are two dots and the probability of nuclear spin-flip process in each of the dots is 50%. We estimate $f_T \approx 0.05$. We then find that D in Fig. (1) is given as $D = f_T A^2 \Gamma/N^2\hbar(\Delta_{ST}^2 + \Gamma^2)$.

The quadrupole interaction of As nuclei is much stronger than that of Ga [$\nu_{As} \approx 2.5$ MHz is in the same order as the Zeeman energy $\omega_{As} \approx 5.9$ MHz (at 0.8 Tesla)] and leads to a complete mixing of the spin eigenstates, so that the perturbative wavefunctions [Eq. (2)] for Ga nuclei have to be replaced with numerical solutions:

$$|\phi_i\rangle = \sum_m u_{im}|m\rangle. \qquad (10)$$

The expectation value of $\hat{I}_z$ of these state is $\langle \phi_i|\hat{I}_z|\phi_i\rangle = \sum_m |u_{im}|^2 m$. For example, we find that when $\alpha \equiv \nu_{As}/\omega_{As} = 0.3$, $\langle \phi_i|\hat{I}_z|\phi_i\rangle = 1.419$, 0.558, -0.528 and -1.450 for $i = 3/2$, 1/2, -1/2 and -3/2 respectively. All the matrix elements of $\hat{I}_z$ and $\hat{I}_-$ can be easily obtained from Eq. (10), and the transition rates can be calculated numerically using Eq. (6) and (9). One should keep in mind that the illustration of the various transitions in Fig. (1) is only applicable to the perturbative analysis for Ga nuclei, while for As nuclei all transitions are possible. However the concept of polarization (D) and depolarization (F and G) processes illustrated in Fig. (1) should still be valid.

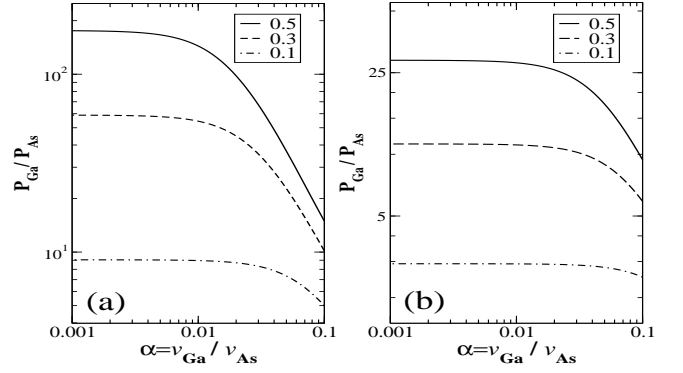FIG. 2: Ratio of Ga and As nuclei polarization as a function of the relative quadrupole interaction strength α ($= \nu_{Ga}/\nu_{As}$) for $\Gamma = 10$ μeV (panel a) and $\Gamma = 30$ μeV (panel b). Γ is the width of level broadening due to electron tunneling between the lead and the dot. The three curves in each graph represent three different As quadrupole coupling strengths $\nu_{As}/\omega_{As} = 0.1$, 0.3 and 0.5. θ has chosen to be 45 degrees for both As and Ga nuclei.

After obtaining all the transition rates W^z and W^- we can proceed to construct the master equations [13]

$$\frac{dp_i}{dt} = \sum_{j\neq i} W_{j,i}p_j - \sum_{j\neq i} W_{i,j}p_i, \qquad (11)$$

where $W_{i,j} = W_{i,j}^z + W_{i,j}^-$, and p_i is the probability of nuclear spins in state $|\phi_i\rangle$. In Eq. (11) we have neglected the spin-lattice relaxation, which is very weak at liquid Helium temperature. Here we can use the spin temperature approximation because the nuclear spin build-up time (≈ 100 s) [6] is much longer than the transverse spin relaxation T_2 ($\approx 10^{-4}$ s). We solve for the steady-state polarization: $dp_i/dt = 0$. These equations are highly nonlinear, as the equilibrium polarizations depend on the transition rates, while to calculate the transition rates W^- one must know the effective nuclear magnetic field, which itself depends on the nuclear spin polarization. We solve the set of equations self-consistently. The average nuclear polarization is $P = \sum_i p_i\langle \phi_i|\hat{I}_z|\phi_i\rangle = \langle \hat{I}_z\rangle/I$. We can then calculate the effective nuclear magnetic fields B_N with Eq. (8).

Figure 2 shows the calculated ratio of Ga and As nuclei polarization as a function of α, which is taken as a free parameter in our calculations. We consider As quadrupole effects with three different strengths. $\nu_{As}/\omega_{As} = 0.3$ corresponds to $\nu_{As} \approx 1.8$ MHz. In this case the Ga nuclear polarization is two orders of magnitude greater than that of As nuclei when $\alpha \approx 0.01$. This difference can qualitatively explain the experimental observation that the As NMR signal is missing in measuring the current oscillation period as a function of the AC magnetic field frequency. We also find the ratio is very sensitive to electron level width Γ. Figure 2 shows that the polarization
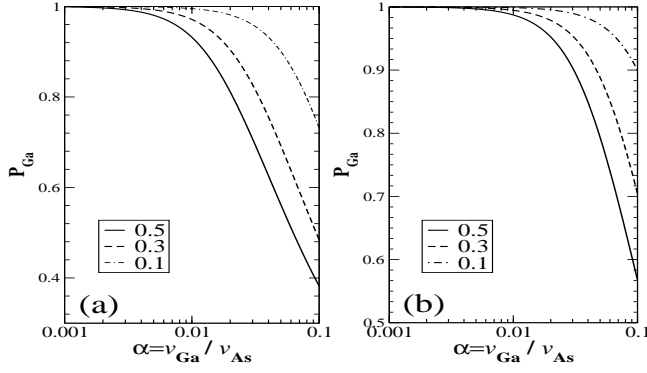
FIG. 3: Ga nuclear spin polarization as a function of the relative quadrupole interaction strength α for $\Gamma = 10$ μeV (panel a) and $\Gamma = 30$ μeV (panel b). All the parameters are the same as those used in Fig. 2.

ratio in the case of $\Gamma = 30$ μeV is roughly $\frac{1}{5}$ of that in the case of $\Gamma = 10$ μeV. Since $D \propto \Gamma/(\Delta_{ST}^2 + \Gamma^2)$, the triplet to singlet transition rate decreases much faster when Γ is smaller, resulting in larger polarization ratio.

Figure 3 shows how Ga nuclear spin polarization changes with the relative quadrupole coupling strength α. It is obvious from the two panels that P_{Ga} increases rapidly as α decreases from 0.1 to 0.01. The Ga polarization saturates to unity for smaller α, and larger Γ leads to larger Ga polarization.

The physical picture of the master equation calculation is actually quite straightforward. Both Ga and As have a polarization rate and a depolarization rate. The As nuclear spin depolarization rate is much larger than that of Ga due to the quadrupole coupling. Thus there exists a regime of polarization rate where nuclear spin polarization can be built up in Ga but not in As.

In our study As depolarization rates are greatly enhanced because the energy levels of the dots are broadened through tunnel coupling to the leads, and that the singlet-triplet transition becomes off resonance once the polarization of Ga nuclei is established. In previous experiments concerning dynamical nuclear polarization, optical pumping creates a constant polarization rate for the nuclei with the help of hyperfine interaction; and the relaxation process due to carrier recombination overshadows the effect of quadrupole interaction, so that As polarization can still be built up [1].

In conclusion, we have studied the dynamical nuclear polarization mechanism in spin-blockade double-dot system. Specifically, we calculate the hyperfine induced polarization rates and depolarization rates among the eigenstates of the nuclear spins. We show that the average spin polarization in steady state of different nuclear species (Ga and As) could differ by two orders of magnitude due to static quadrupole interaction induced by lattice distortions. Our results can thus be used to explain the recent NMR experiments conducted in such system. Our calculation suggests both caution and promise. Doping is widely used in semiconductors to tailor electronic properties. However, as we point out in this Letter, there can be unexpected side effects in material properties, such as increased nuclear spin relaxation in the present case. Conversely, controlled doping can also be used to differentiate parts of a system (such as Ga and As nuclei here), so that selective operations become possible. How to utilize the additional control provided by doping while overcoming its negative effects can be critical to future quantum information processors and/or spintronic devices.

[1] F. Meier, and B.P. Zakharchenya, *Optical Orientation* (North-Holland, Amsterdam, 1984).
[2] D. Gammon *et al.*, Science **277**, 85 (1997).
[3] R.K. Kawakami *et al.*, Science **294**, 131 (2001).
[4] A.V. Khaetskii and Yu.V. Nazarov, Phys. Rev. B **64**, 125316 (2001); J. Schliemann, A.V. Khaetskii, and D. Loss, *ibid.* **66**, 245303 (2002); R. de Sousa and S. Das Sarma, *ibid.* **67**, 033301 (2003); *ibid.* **68**, 115322 (2003).
[5] A. Imamoḡlu, E. Knill, L. Tian, and P. Zoller, Phy. Rev. Lett. **91**, 017402 (2003).
[6] K. Ono and S. Tarucha, cond-mat/0309062.
[7] K. Ono, D.G. Austing, Y. Tokura, and S. Tarucha, Science **297**, 1313 (2002).
[8] L.P. Kouwenhoven, D.G. Austing, and S. Tarucha, Rep. Prog. Phys. **64**, 701 (2001).
[9] E.H. Rhoderick, J. Phys. Chem. Solids **8**, 498 (1958).
[10] W.E. Carlos, S.G. Bishop, and D.J. Treacy, Phy. Rev. B **43**, 12512 (1991); Appl. Phy. Lett. **49**, 528 (1986).
[11] The crystal structure of *pure* GaAs is zinc-blende with cubic symmetry. No noticeable field gradient is expected due to this symmetry.
[12] J.V. Kranendonk, Physica **20**, 781 (1954).
[13] A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, London, 1961); C.P. Slichter, *Principles of Magnetic Resonance* (Spinger-Verlag, Berlin, 1996).
[14] D. Paget, G. Lampel, B. Sapoval, and V.I. Safarov, Phys. Rev. B **15**, 5780 (1977).
[15] Y.B. Lyanda-Geller, I.L. Aleiner, and B.L. Altshuler, Phys. Rev. Lett. **89**, 107602 (2002).
[16] We have assumed that the electron g-factor is negative and the triplet is the ground state at zero magnetic field. One could equivalently discuss the other triplet state $|1,1\rangle = |\uparrow\rangle|\uparrow\rangle$ if the singlet is the ground state when there is no magnetic field [17].
[17] T. Inoshita, K. Ono, and S. Tarucha, J. Phys. Soc. Jpn. **72** 183 (2003).